LANL-SuperPower CRADA Strategic Research

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FY 2005 plans

- Consult with our industrial partner(s) to address strategies of mutual interest for increasing performance and reducing costs.
- Modify IBAD assist gun to expand deposition zone length to improve process efficiency. *Goal: Double length of deposition zone*.
- Improve IBAD MgO texture by reducing divergence of SuperPower's ion-assist gun. *Goal: Routinely obtain $\Delta\Phi \leq 5°$ FWHM*.
- Expand upon our data of how YBCO superconducting properties (T_c, J_c) are affected by transition metal impurities. *Goal: Determine tolerance limits for substrate elements in YBCO films*.

Many current and near-term production related issues were supported throughout the past year

- Electropolishing of alternate substrate metal alloys of interest to SuperPower

- Analyses (SEM, FIB, TEM, STEM, EDS, RHEED, ISS, XRD, RBS, PIXE, AFM) of film stack(s) for thickness verification, interfacial integrity, possible interdiffusion of layers, stoichiometry, impurity information, etc.

- LANL depositions on SuperPower IBAD and buffer layers to verify integrity/texture of different layers and/or answer production/quality related issues

- Software for more accurate and rapid interpretation of local I_c values from magnetic imaging data

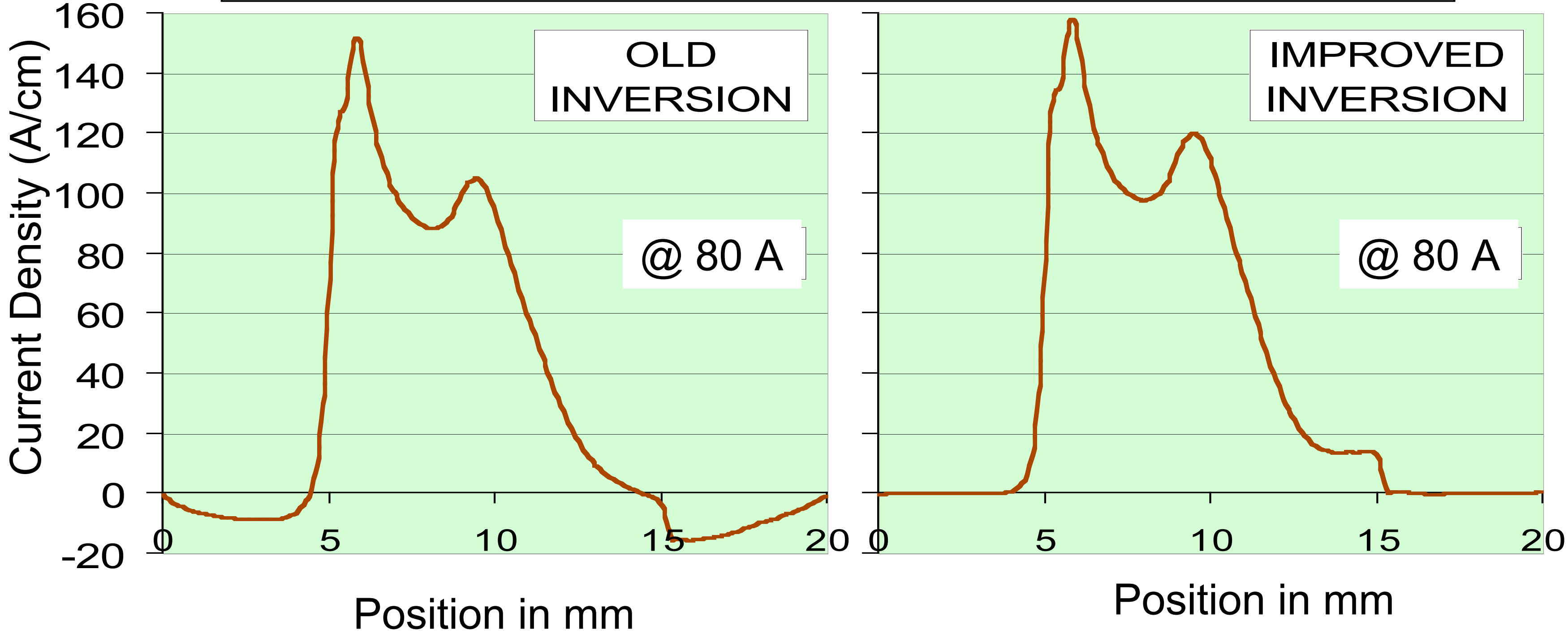
- Successfully demonstrated YBCO-Y_2O_3 multilayers for expected compatibility with SuperPower MOCVD YBCO processing

- $J_c(T,H,\theta)$ measurements - understand and improve vortex pinning

- Equipment loans (emergency and long-term) on as-needed bases

New algorithm treats transport currents in magnetic imaging data more accurately than previous methods*

Linear current density distribution in tape cross-section



- **New software ~ 10 X faster than previous inversion algorithms**
- **Amenable to implementation in on-line reel-to-reel measuring system**

*H. Grube, *2005 APS March Meeting*. **Los Angeles CA**

Buffer layers for IBAD MgO

Buffer layers optimized at LANL for IBAD MgO template (YBCO/*STO/homo-epi MgO*/ IBAD MgO) result in excellent I_c, J_c, but MgO and STO buffers are incompatible with SuperPower's use by high-rate reactive sputter deposition processing

- Sr-Ti: cannot be fabricated into a metal alloy target
- MgO: high Mg vapor pressure results in metal evaporating from deposition surface before reacting with background O_2 (thickness control issue)

Decided to focus on MeZrO and MeHfO buffers

- T_m(hafnates, zirconates) > T_m(titanates) > T_m(niobates)
- Very refractory oxides tend to chemical inertness with YBCO (e.g. $BaZrO_3$) (possible advantage for high-rate, high-temperature YBCO deposition)

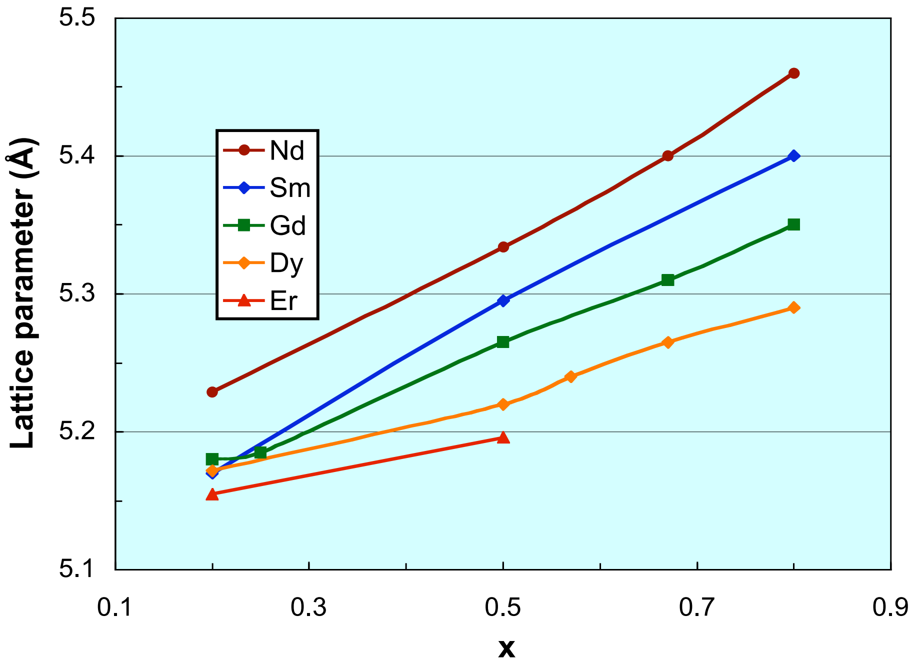
X Like Sr-Ti, Ba-Zr cannot be fabricated as an alloy target

RE-Zr and RE-Hf satisfy several Hume-Rothery conditions for alloy formation

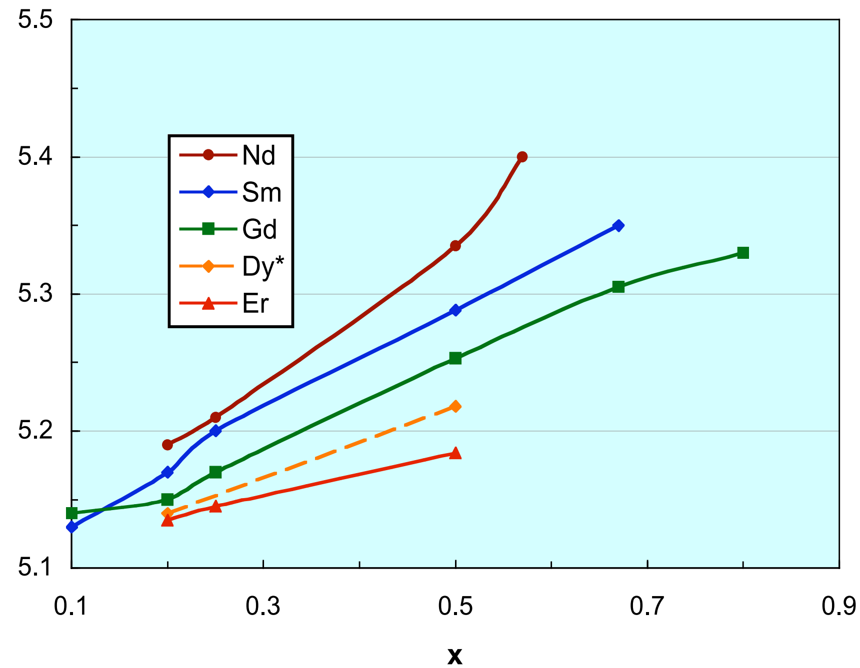
- Focused on least expensive RE elements (e.g. Nd, Sm, Gd, Dy, Er)
- These form stable alloys with Zr, Hf

REZrO and REHfO form cubic phases over a wide range of stoichiometries

Cubic phases of $RE_xZr_{1-x}O_y$ compounds as a function of RE atomic fraction



Cubic phases of $RE_xHf_{1-x}O_y$ compounds as a function of RE atomic fraction



- **If film stoichiometry drifts during long length processing, cubic phase will be retained**

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* $Dy_{0.2}Hf_{0.8}O_y$ lattice parameter estimated

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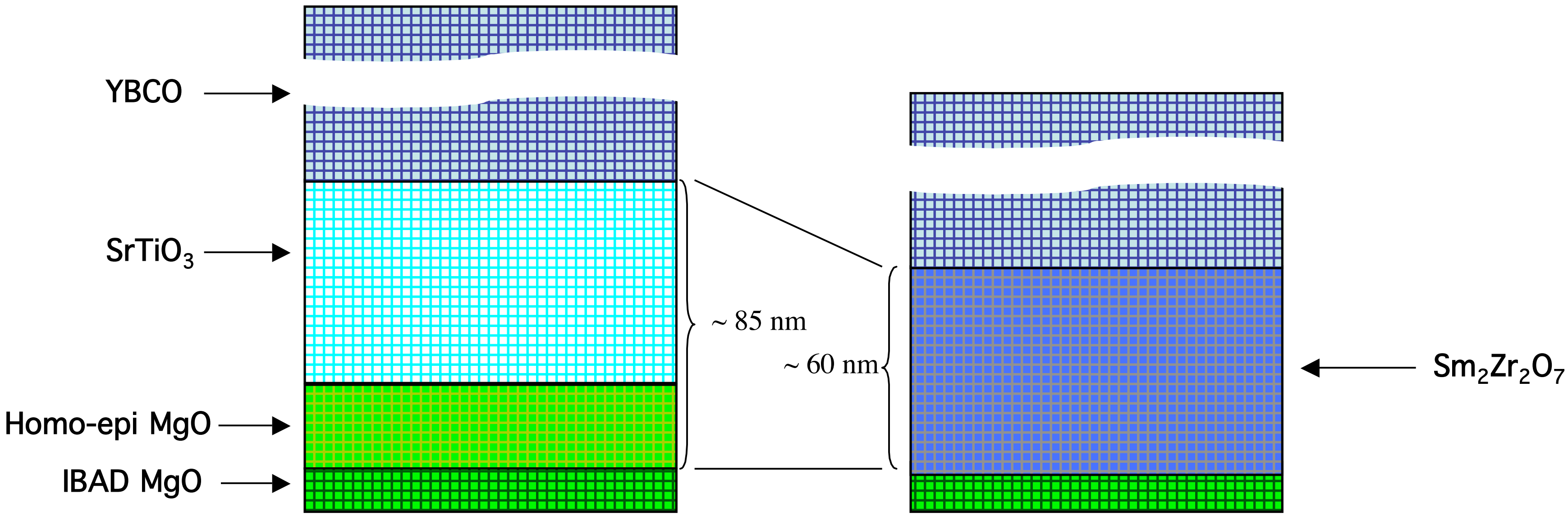
Two buffers replaced by one, and thickness reduced - initial PLD YBCO results very promising for use by reactive sputtering



IBAD MgO: $\Delta\phi$ = 6.2°, $\Delta\omega$ = 2.3°

$Sm_2Zr_2O_7$: $\Delta\phi$ = 4.4°, $\Delta\omega$ = 2.5°

YBCO: $\Delta\phi$ = 2.7°, $\Delta\omega$ = 1.1°

T_c = 89 K, thickness = 1.5μm,

J_c = 2.6 MA/cm² ⇨ I_c = 390 A/cm-width

Several effects occur simultaneously during growth of films via IBAD texturing…

1. Anisotropy of ion induced damage

   W. Ensinger, *Nucl. Inst. and Meth.* B **106** (1995) 142.
   L. Dong, *Appl. Phys. Lett.* **75** (1999) 584.*

2. Anisotropy of sputter yield

   L. S. Yu, *Appl. Phys. Lett.* **47** (1985) 932.
   L. Dong. *J. Appl. Phys.* **89** (2001) 4105.*
   R. Huhne, *J. Appl. Phys.* **47** (2001) 1035.*

3. Anisotropy of defect annihilation

   B. P. Uberuaga, Phys. Rev. Lett. **92** (2004) 115505.*
   I. Usov, *Nucl. Inst. and Meth. B* (submitted)*

4. Anisotropy of ion channeling

   J. Lindhard, *K. Dan. Mat. Fys. Medd.* **34(14)** (1965) 1.

*Papers referring either to theoretical or experimental studies of MgO

…prompting a study of ion induced damage anisotropy while attempting to suppress other effects*

MgO single crystals of (100), (110) and (111) orientations were implanted with 100 keV Ar^+ ions at varying doses and implantation temperatures.

- At 100 keV sputtering is negligible (2)
- Damage centered at depth ~ 100 nm below crystal surface (3)
- Implants were performed 7° from normal to minimize channeling (4)
- Larger defect clusters are expected at this energy vs. ~ 1 keV range used during IBAD**

* I. Usov, *Nucl. Inst. and Meth. B* (In press)
** B. P. Uberuaga, *Phys. Rev. Lett.* **92** (2004) 115505.

Rutherford backscattering spectrometry, combined with channeling, gives information on the concentration and type of defects*

The damage criteria employed in our study are the following**:

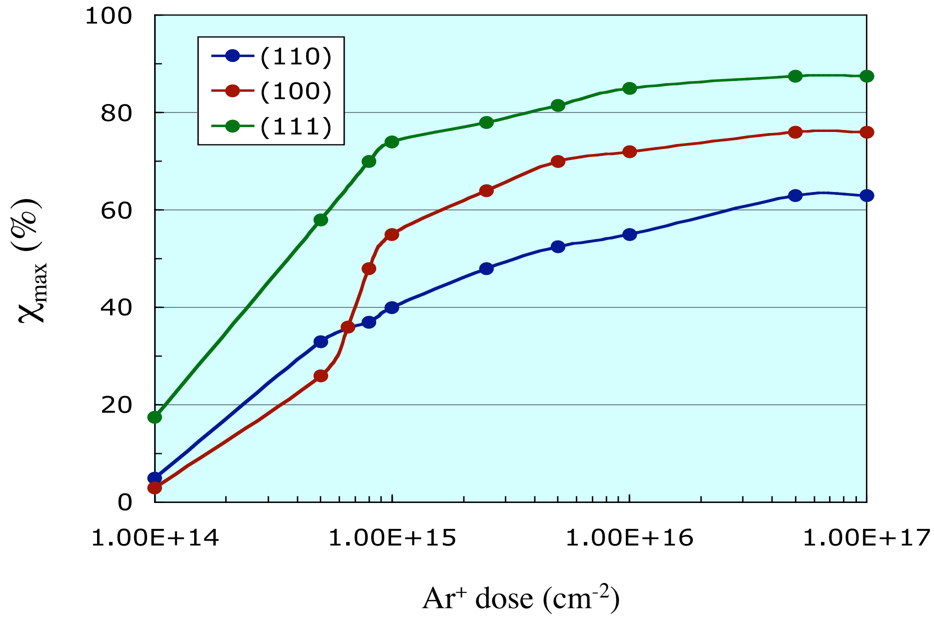
1. χ_{min} is dominated by lattice distortions surrounding extended defects

2. N_D represents the number of isolated atoms displaced from lattice sites (interstitials)

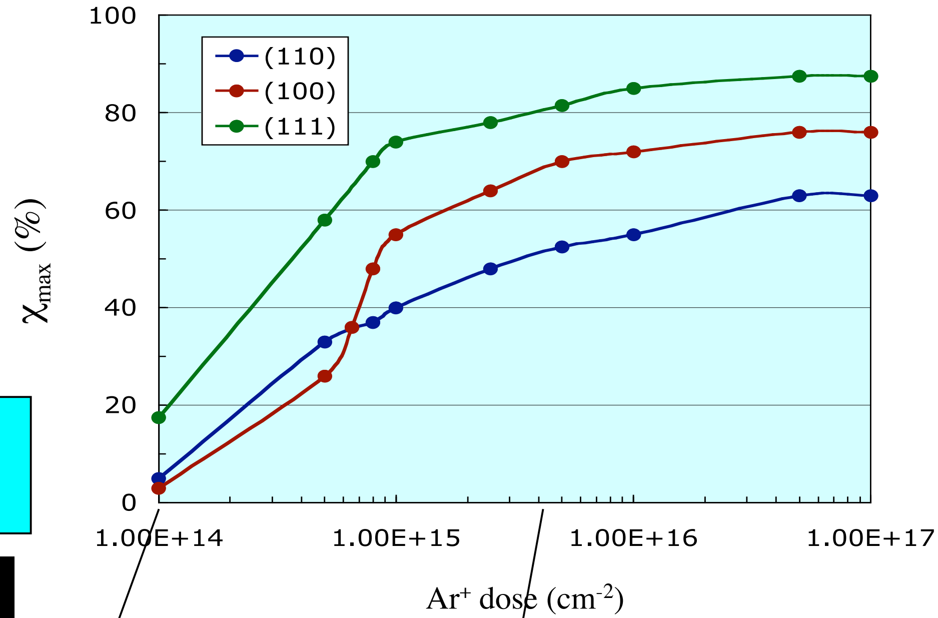
3. χ_{max} is determined by combining both of above defect types

* J.S Williams, in *Ion Beams for Materials Analysis* **82** (Academic Press,1989) p.261
** I. Usov, *Nucl. Inst. and Meth. B* (submitted)

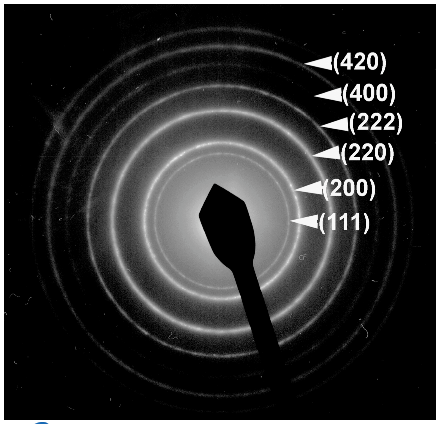
Review: last year Ar^+ damage anisotropy of MgO crystals indicated a possible explanation for IBAD MgO texturing mechanism…



…but a question arises as to what occurs if the assist beam current density is too low

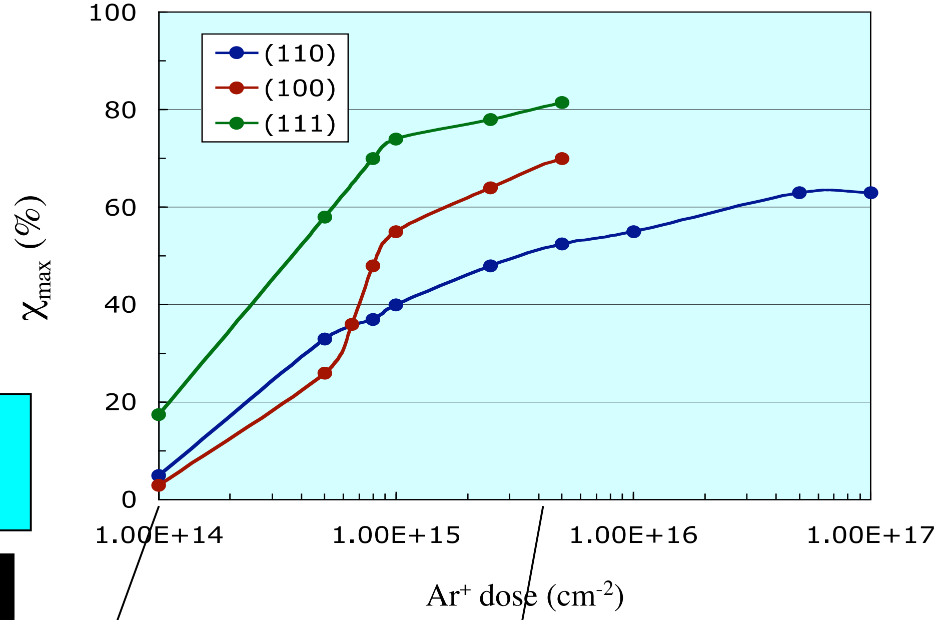


Plan view SAD pattern of non-IBAD MgO film on a-Si_3N_4 exhibits random orientation.



If ion dose is too low, all MgO orientations "survive" bombardment - result is randomly oriented film

As dose increases, at some point, all orientations but the (110) are removed by sputtering …



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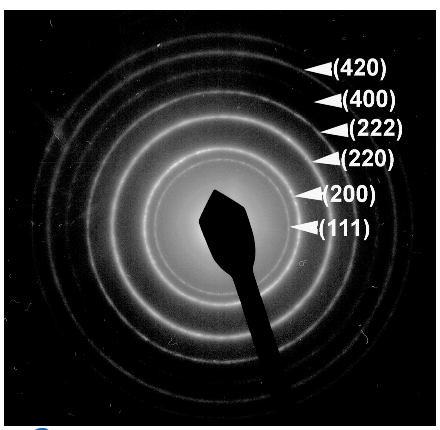

If ion dose is too low, all MgO orientations "survive" bombardment - result is randomly oriented film

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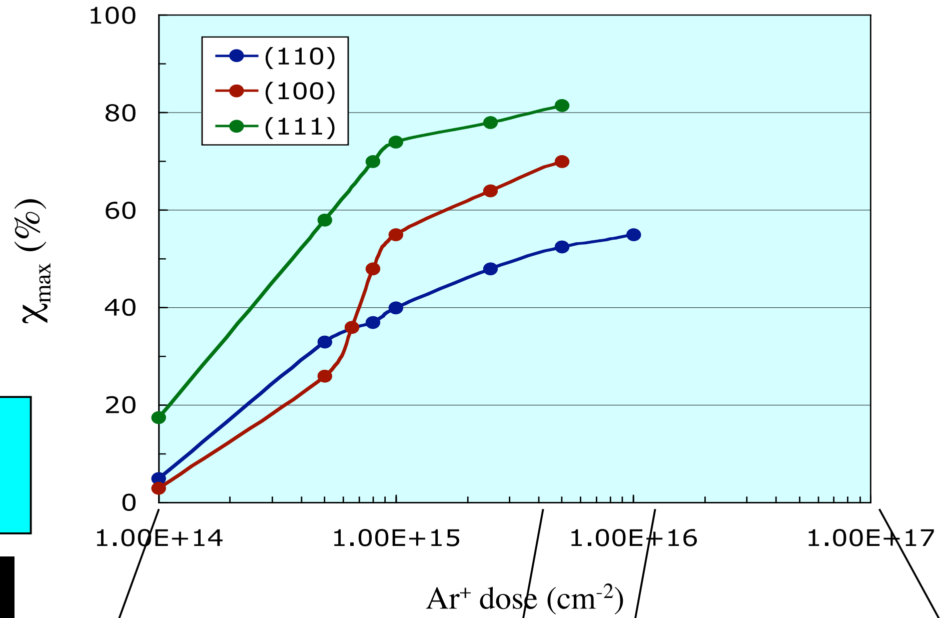
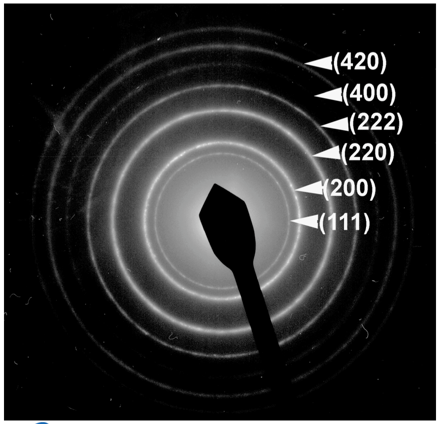
…and at even higher doses, the (110) oriented grains are also removed



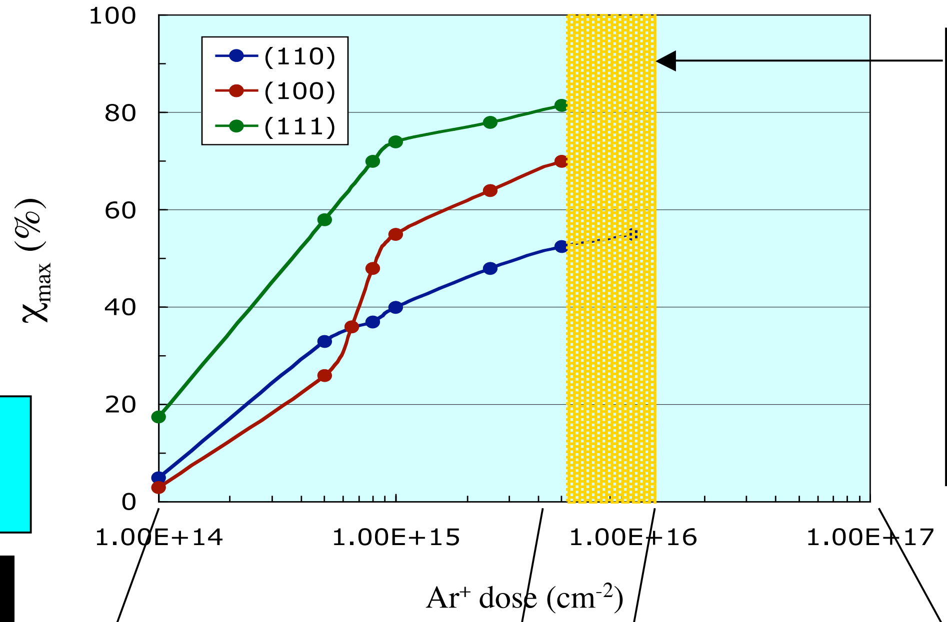
Plan view SAD pattern of non-IBAD MgO film on a-Si_3N_4 exhibits random orientation.



If ion dose is too low, all MgO orientations "survive" bombardment - result is randomly oriented film

If ion dose is too high, the remaining MgO is completely removed by sputtering

For optimum assist-ion/atom ratios, textured material is grown



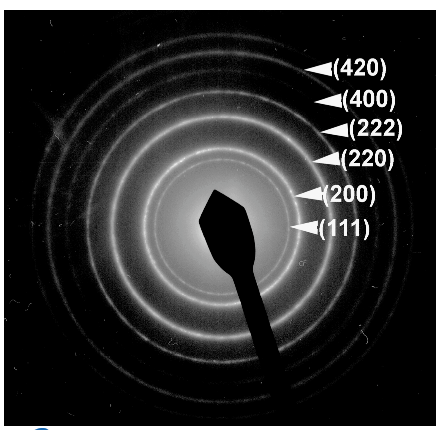
Dose range used by various institutions during IBAD MgO processing

APL **71** p.2955 (1997)
IEEE TAS **9** p.1964 (1999)
JAP **90** p.1035 (2001)
APL **80** p.3388 (2002)
JAP **93** p.205 (2003)

If ion dose is too low, all MgO orientations “survive” bombardment - result is randomly oriented film

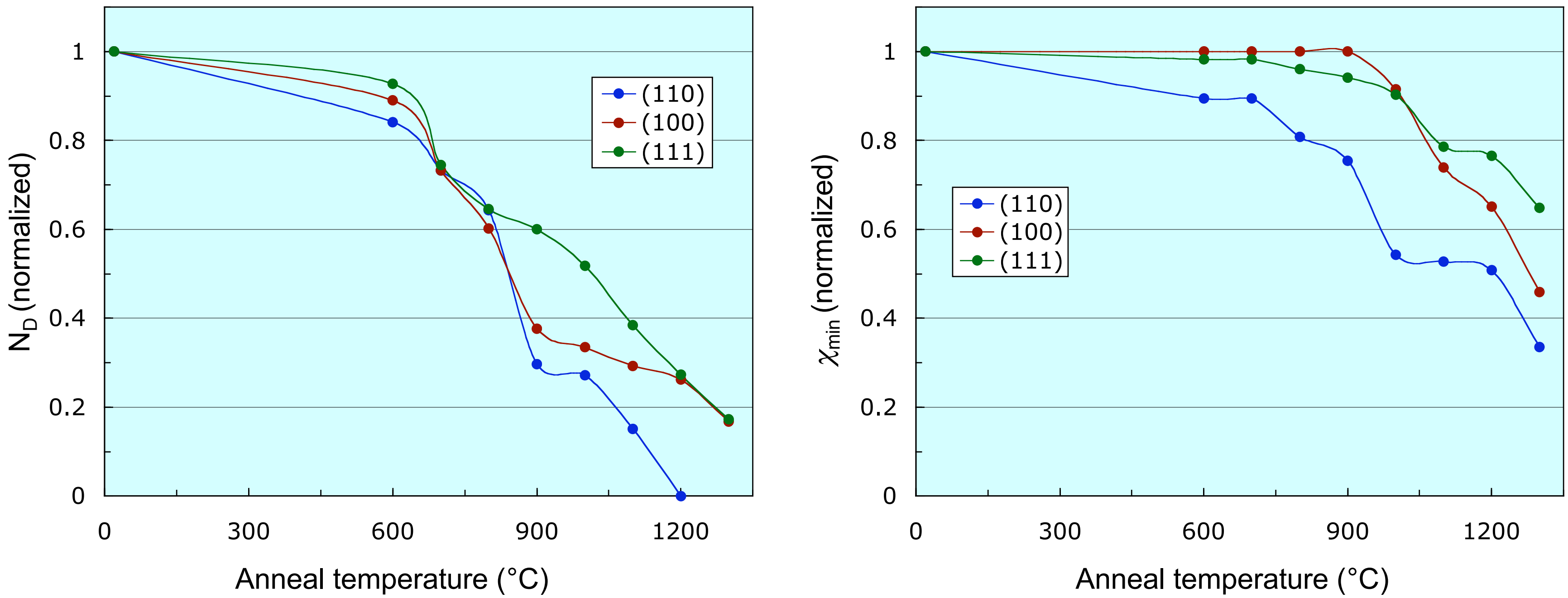
If ion dose is too high, the remaining MgO is completely removed by sputtering

Plan view SAD pattern of non-IBAD MgO film on *a*-Si_3N_4 exhibits random orientation.



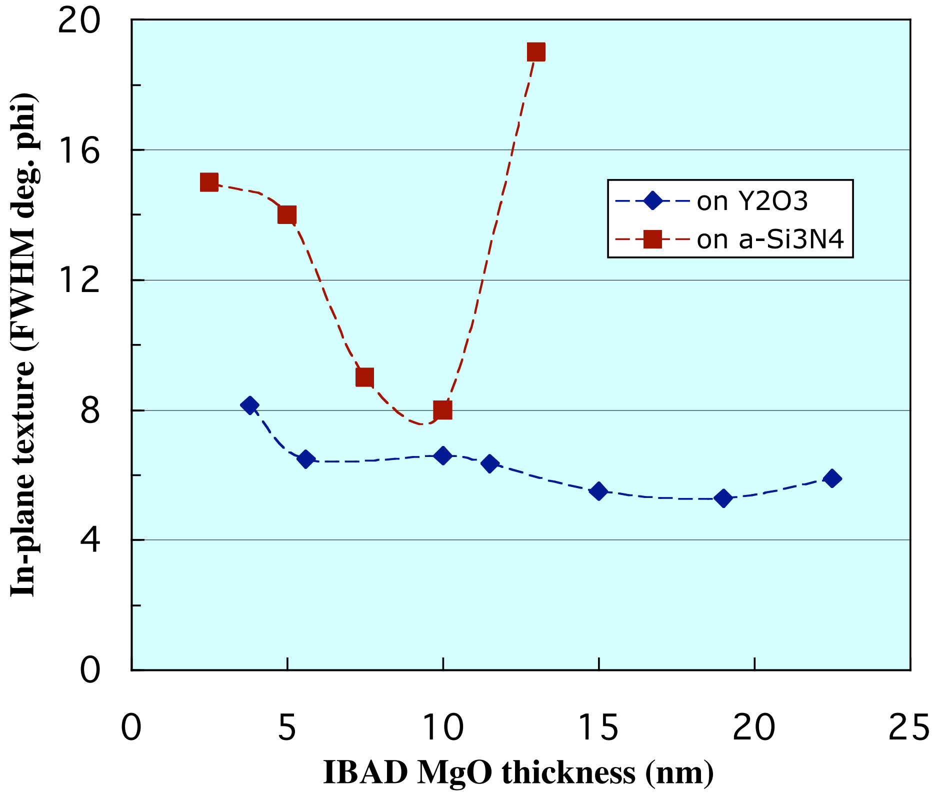
Rapid thermal anneal studies of MgO crystals were initiated to determine how orientation influences damage recovery

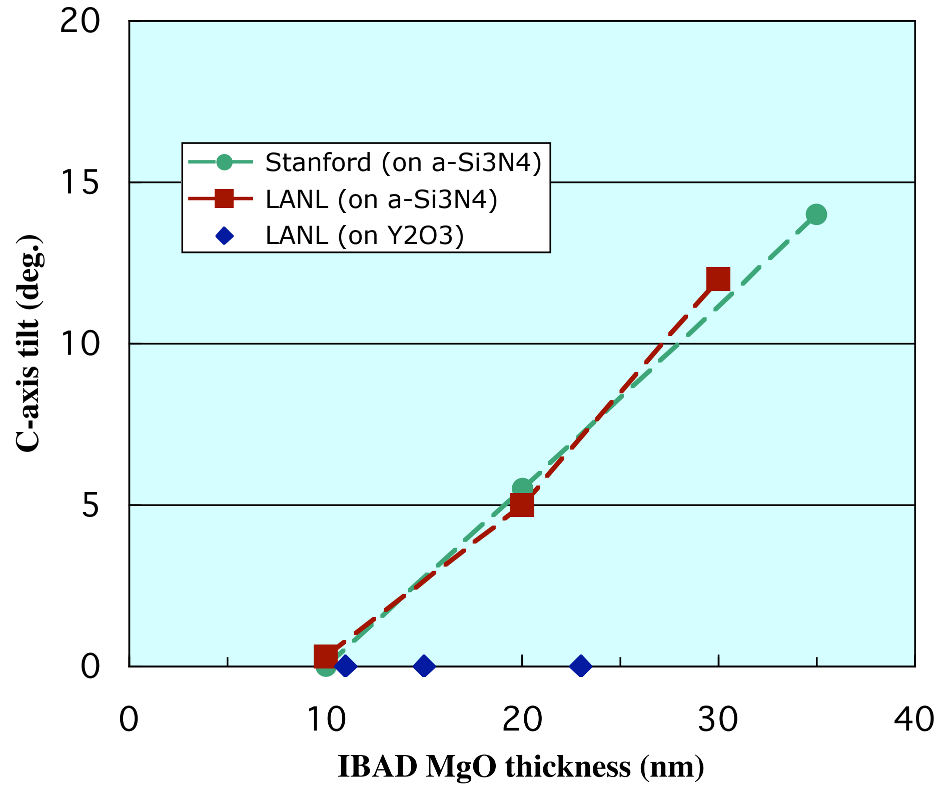
Dependence of N_D & χ_{min} on anneal temperature for MgO single crystals
(Ar^+ dose = $1x10^{15}/cm^2$, anneal time = 3 min.)



- **Annealing of interstitial defects more effective than for extended defects**
- **Amount of recovery greatest for (110) oriented samples - independent of damage criterion**
- **Latter point corroborates theoretical prediction of Uberuaga (Phys. Rev. Lett. 92 (2004) 115505)**

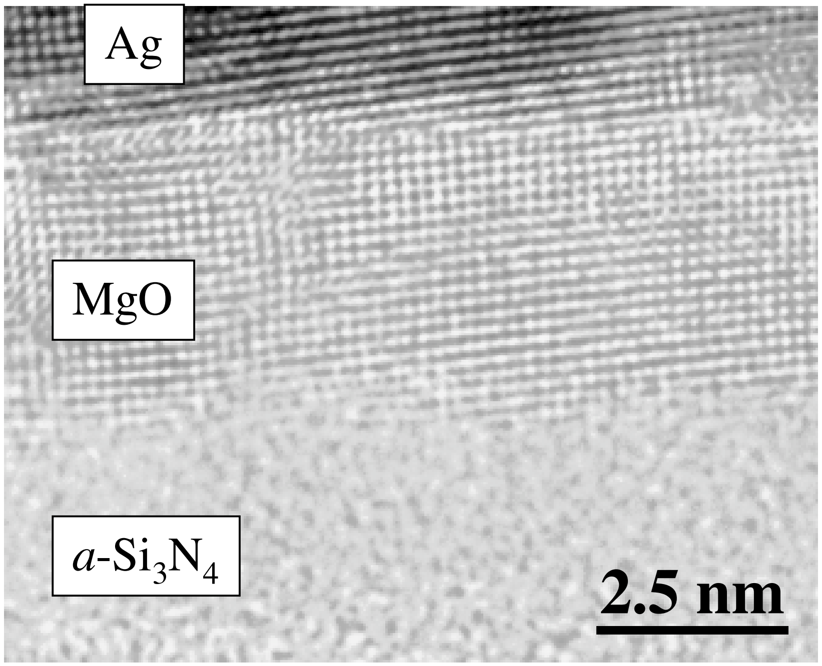
Nucleation layer greatly influences IBAD MgO film growth*

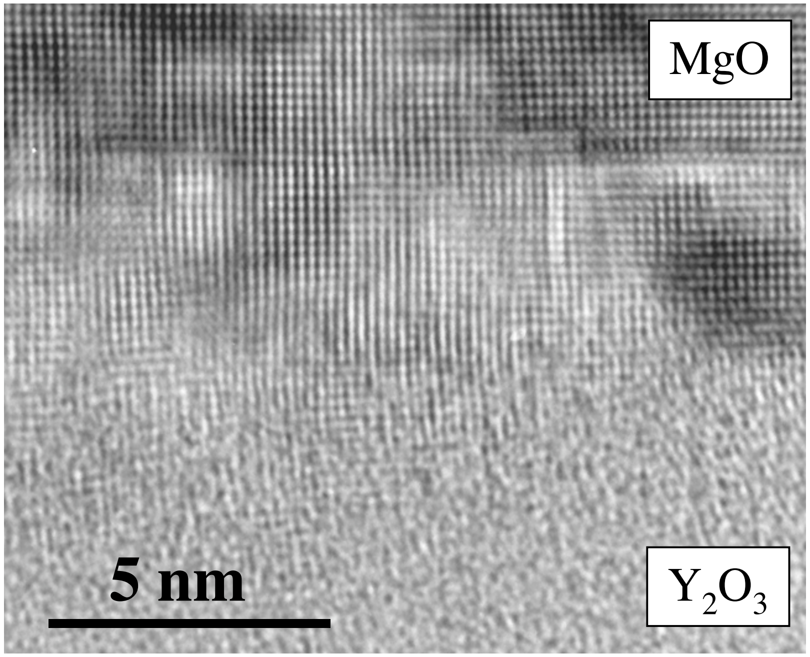




*J.R. Groves, *IEEE Trans Appl. Supercond.* **13** (2003) p. 2651

Comparison of HRTEM cross sections reveals differing growth characteristics for IBAD MgO on the two nucleation layers



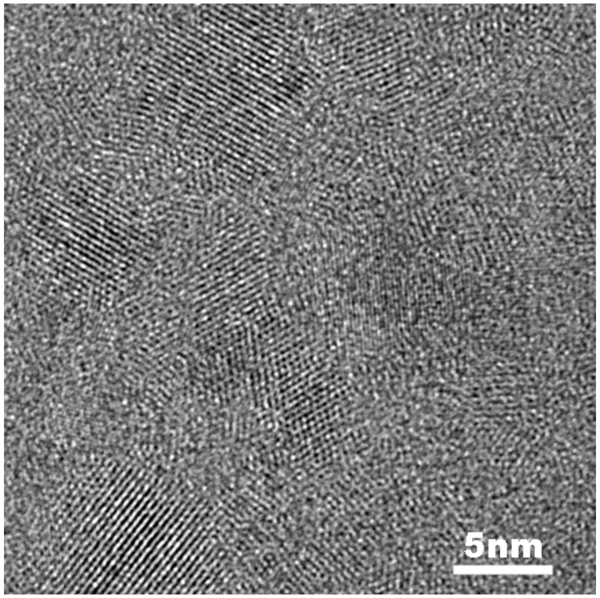


Planar alignment of the layers exhibited within first few monolayers. Slight roughness at the MgO/*a*-Si_3N_4 interface. Grains ~ 10 nm.

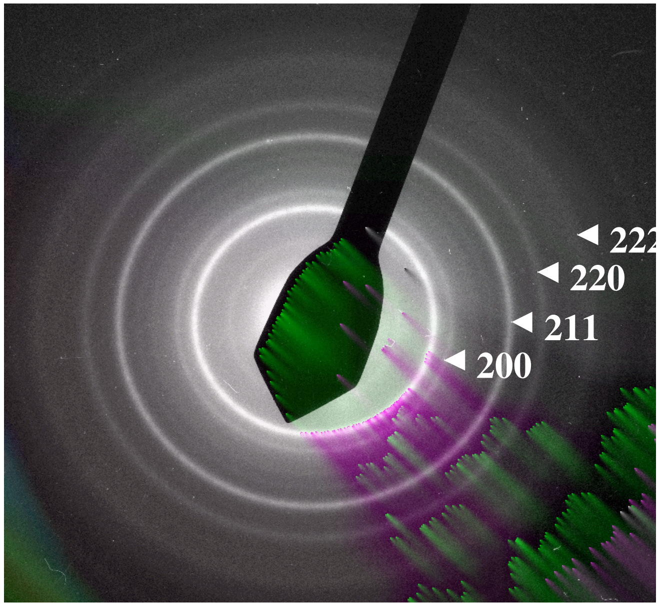
Planar alignment of the layers not exhibited until growth of ~ 20 layers. More roughness at the MgO/Y_2O_3 interface. Grains near interface ~ 2 nm.

HRTEM shows the yttria nucleation layer is nanocrystalline

Plan view of 10 nm thick Y_2O_3 film



SAD pattern of Y_2O_3 nucleation layer also exhibits random orientation



Under IBAD deposition conditions, differing sputter yields of Y_2O_3 and nascent MgO grains are thought to result in increased roughness of interface prior to the transition to (100) oriented MgO. Results in smaller, more numerous grains in early IBAD growth stages relative to the films grown on a-Si_3N_4.

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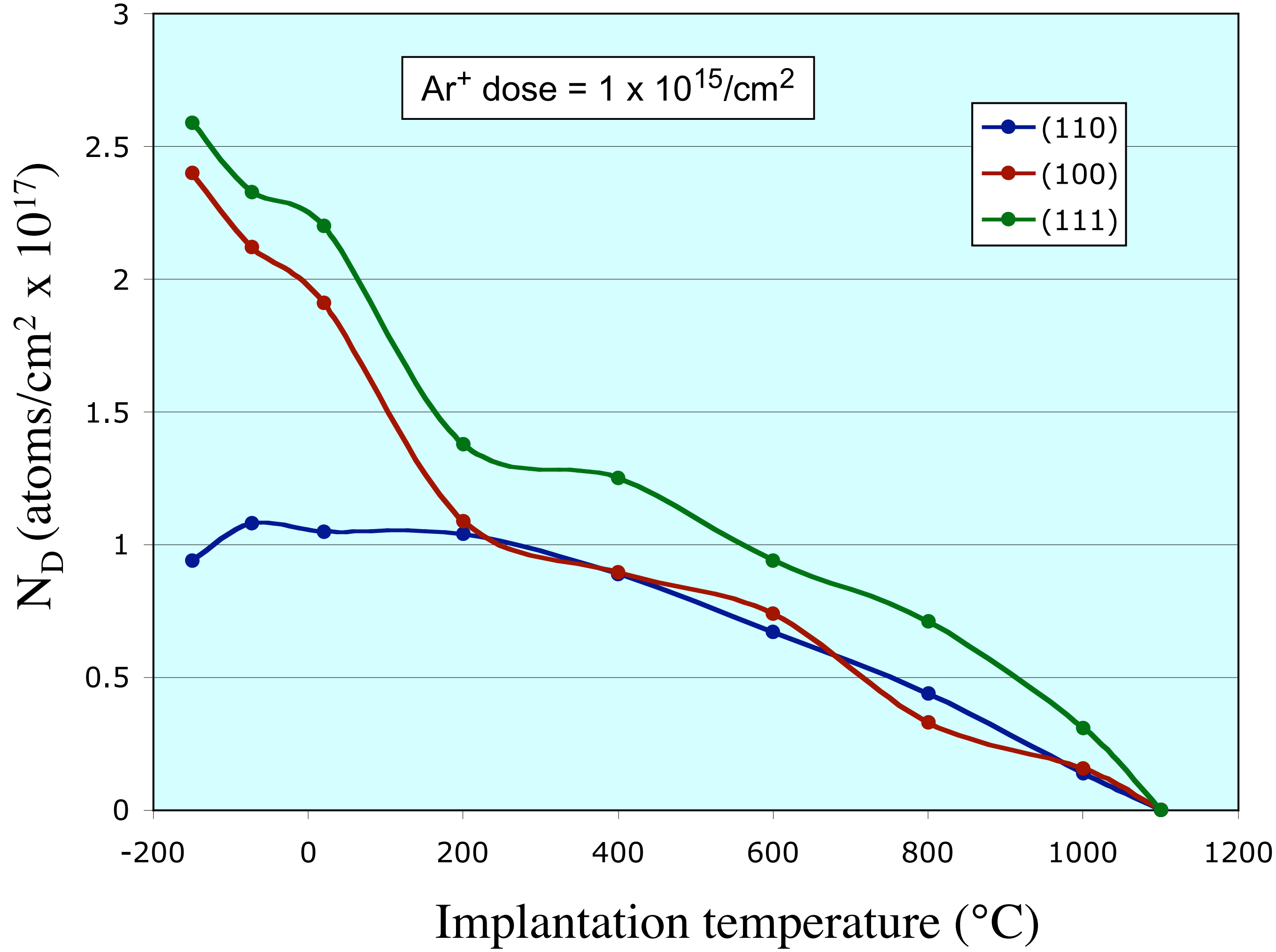
Interpretation of observations

• Buildup of defects is thought to be responsible for c-axis tilt of films on a-Si_3N_4.

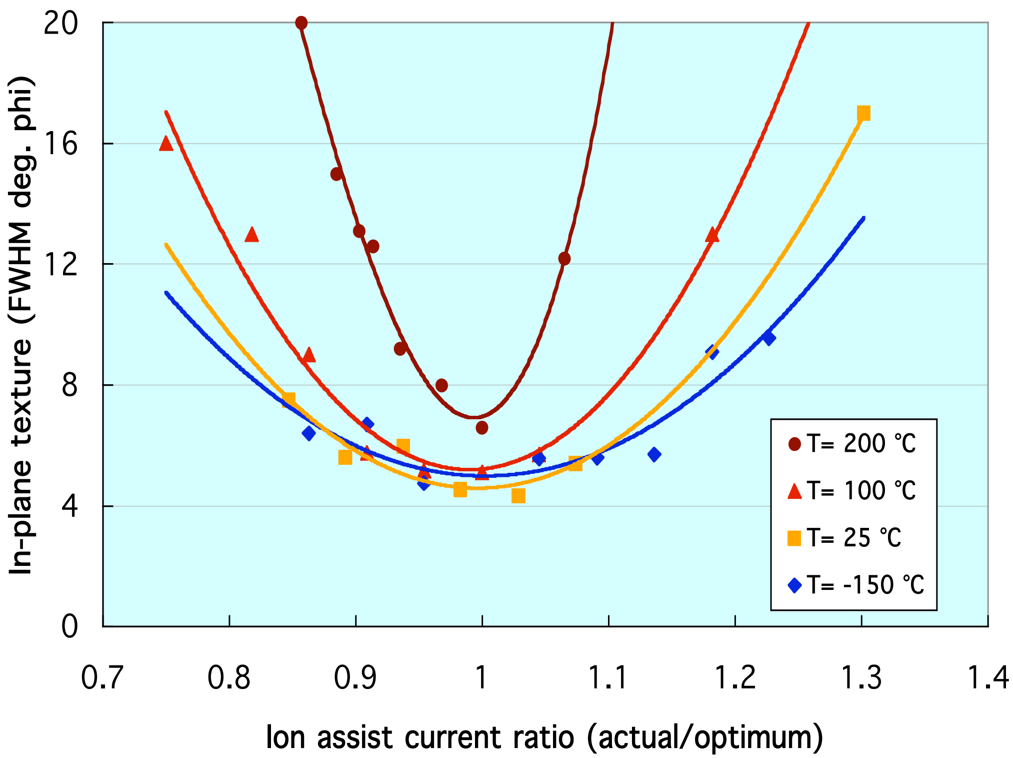
• Defects are mobile and annihilate efficiently on grain boundaries and film surfaces.

• Since the initial IBAD MgO grains on Y_2O_3 are smaller, the probability for annihilation of defects is enhanced. Thus, the onset of the c-axis tilt for the IBAD is delayed when Y_2O_3 is the nucleation layer.

The difference in damage accumulation for the three orientations was greatest at the lowest implant temperatures…



…prompting a study of IBAD MgO texturing vs. deposition temperature



- Stationary substrates were held at fixed temperatures by silver pasting to a copper block during deposition

- The deposition rate was held constant

- Many IBAD samples were made at four temperatures (200, 100, 25 and -150 °C) and at differing ion assist current densities

*S.R. Foltyn, *Appl. Phys. Lett.* **82** (2003) p.4519

- **Non-weak linked YBCO properties demonstrated using IBAD MgO templates of $\Delta\phi \leq 8°$ FWHM***

- **Results indicate cooling metal substrates during reel-to-reel deposition will expand the processing window**

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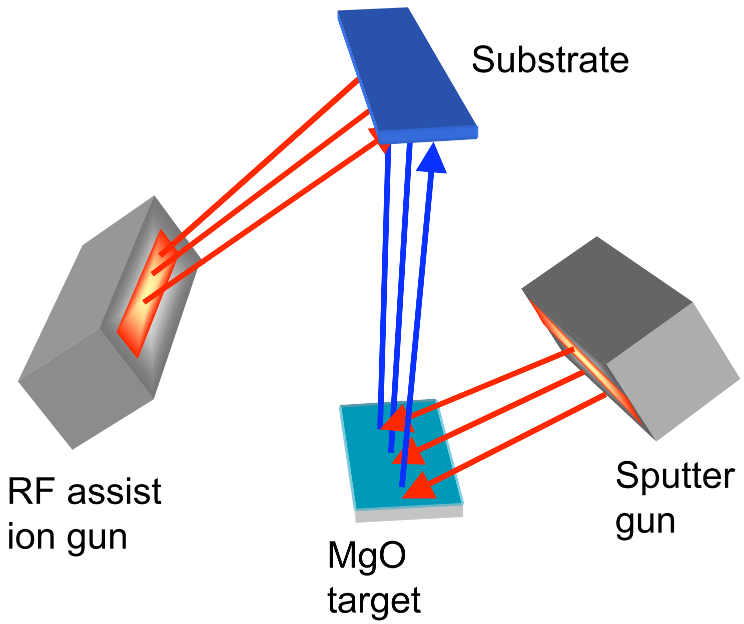
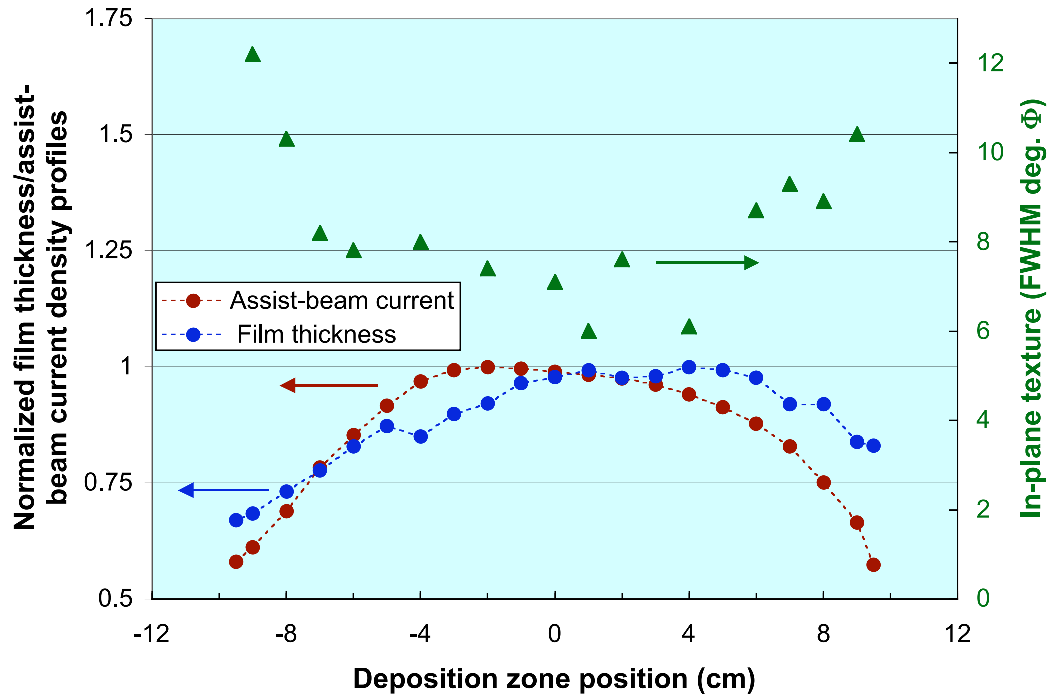
Ion-assist beam current density and film thickness variations along deposition zone affect IBAD MgO film texture*

IBAD deposition schematic

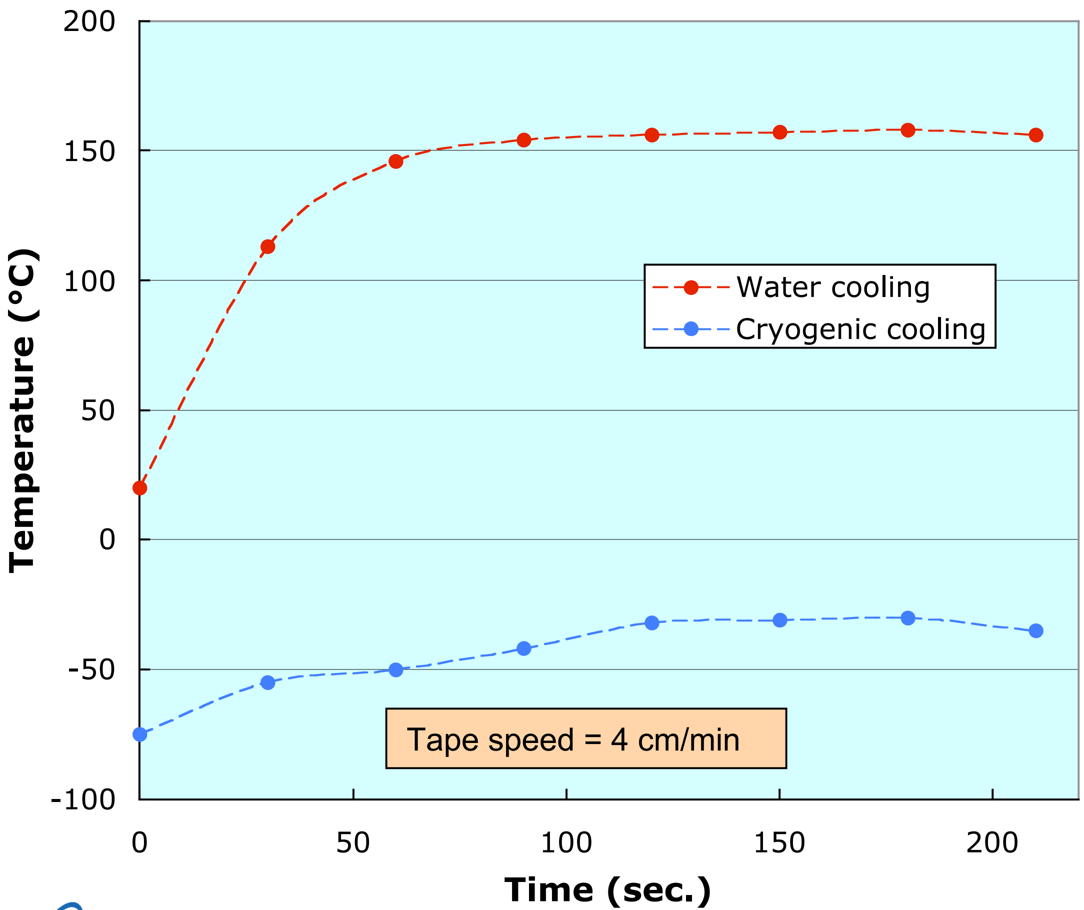




Results corroborate previous $\Delta\phi(T_{sub})$ data of desirability to cool the substrates during reel-to-reel operation before attempting to increasing the deposition zone length

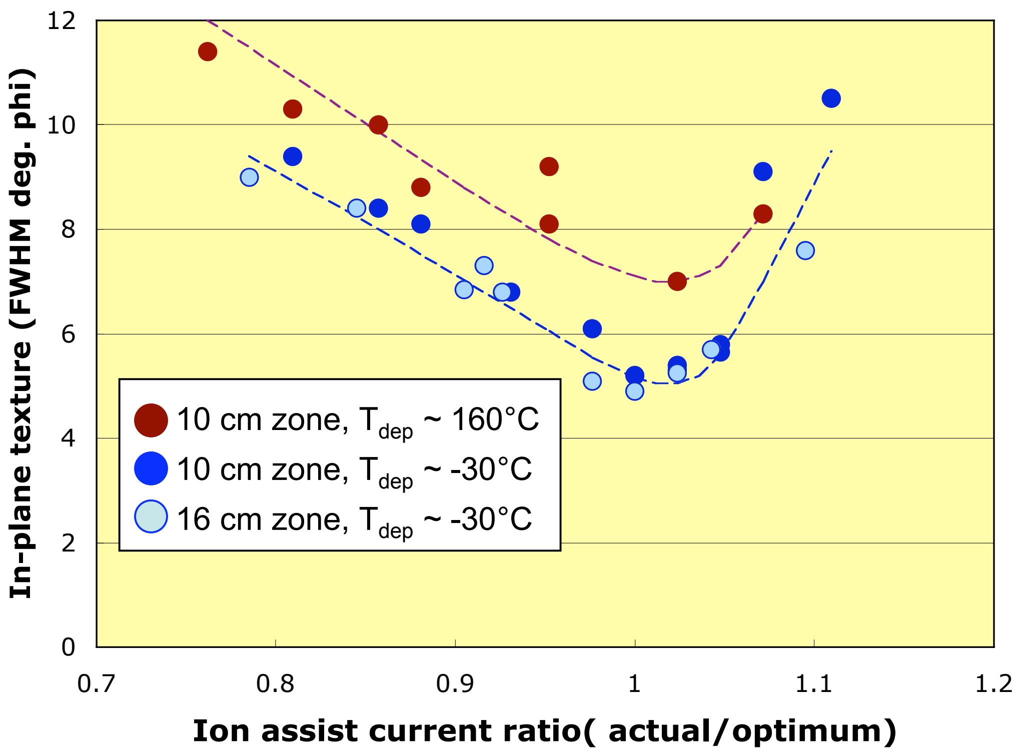
*Films deposited in stationary mode, T_{dep} ~ 150 °C

Moving substrates can be cooled to the lower, more desirable IBAD processing temperatures….



- The substrate cooling block was engineered to enhance contact with moving tapes
- The ion-assist beam (I_B,V_B) was set for SuperPower's IBAD processing conditions
- The tape temperature was measured while moving continuously along the length of the deposition zone

…but will the stationary texture data be repeated for tapes moving though the deposition zone?



To simulate variations in processing parameters for long tape production

•The deposition rate was held constant

• The assist beam current was varied about an optimum value which gave the best texture

Summary

At the lower deposition temperature:

- the IBAD MgO processing window (@ $\Delta\phi \leq 8°$) expands ~ 2X
- overall, $\Delta\phi$ is ~ 2° lower

⇨ A more robust IBAD MgO process is obtained

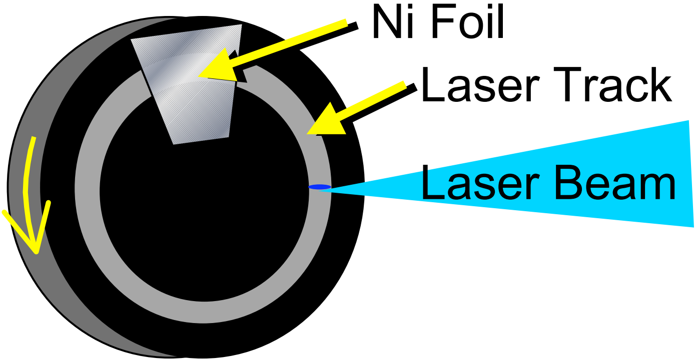
⇨ Manufacturing yield/quality is improved

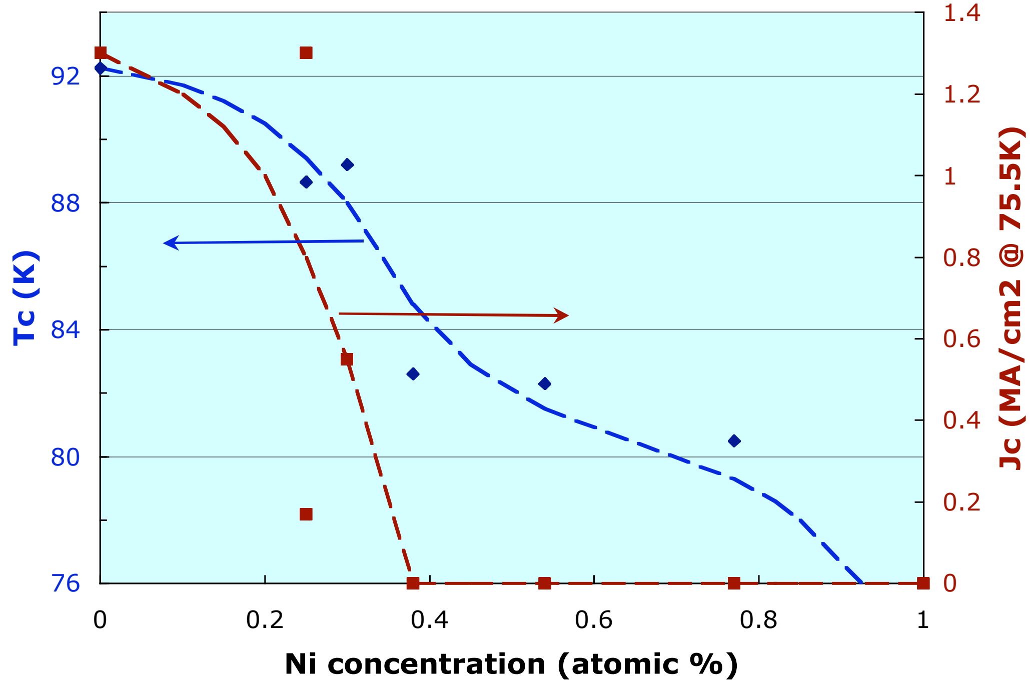
Quantitative tolerance of YBCO for one of the substrate's transition metals?

200 nm PLD YBCO films were deposited on SXAL $SrTiO_3$ substrates. The depositions were performed with or without Ni foil strips mounted on the YBCO target.

T_c and J_c measurements were performed and the Ni concentration was measured using Particle Induced X-ray Emission (PIXE).

PLD deposition schematic





Scoring criterion - Results

1. Replaced two buffer layers with a single layer that is compatible with SuperPower's high rate reactive sputter processing

2. Demonstrated YBCO-Y_2O_3 multilayers compatible with MOCVD processing (I_c = 875A/cm)

3. Developed transport current algorithm - interprets magnetic imaging data faster and more accurately

4. Refined damage anisotropy data - correlating experimental annealing anisotropy data with theoretical prediction

5. Improved our understanding of IBAD MgO texturing mechanisms

6. Refined IBAD MgO $\Delta\phi(T_{sub})$ data for stationary substrates

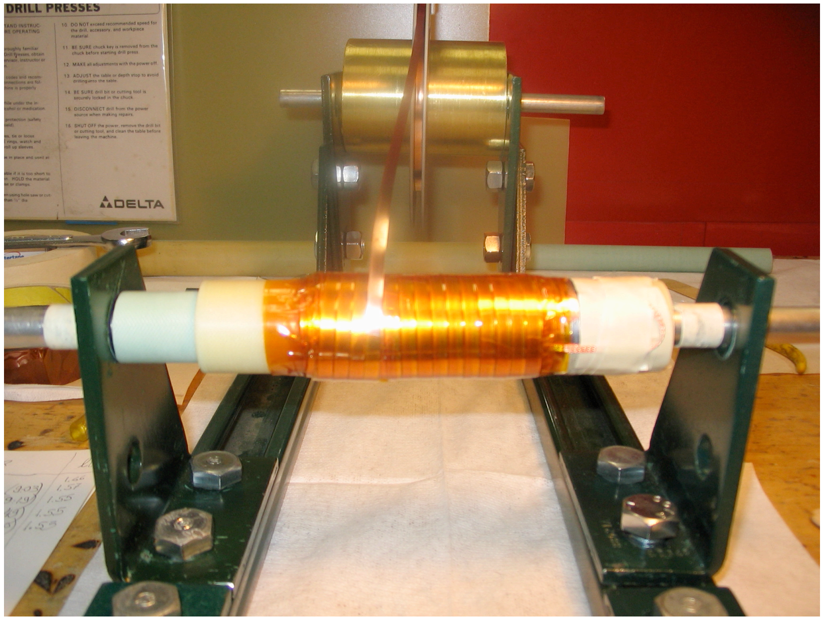
7. Developed electropolishing process for alternate substrate alloy (150 m @ R_a < 1nm

8. Using MOCVD-YBCO CC, LANL hand wound coil - no degradation in I_c=100A/cm @ 75.5 K, L = 9 m, W = 4 mm, Tape thickness ~ 90 µm, Field = 0.12 T @64K

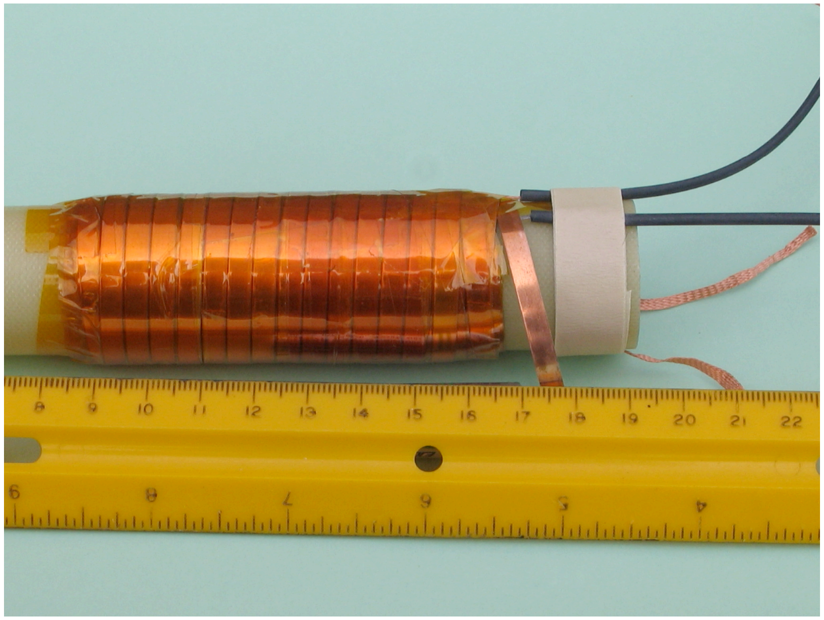
Coil made using SuperPower MOCVD YBCO/IBAD MgO coated conductor

Coil dimensions: Length = 7.5 cm, I.D. = 2.85 cm, O.D. = 3.3 cm, 6 layers, each layer ~ 16 turns, Kapton tape insulation

Hand winding setup



Finished coil



Scoring criterion - Performance

- Consult with our industrial partner to address strategies of mutual interest for increasing performance and reducing costs.
  - Included close interactions, on-site visits, and dedicated teams at SuperPower and LANL. Resulted in the following highlights:
    - SP IBAD MgO ⇨ 100+ m @ 10 m/hour
    - SP IBAD/LANL buffers & YBCO ⇨ 3.0-4.1 MA/cm^2 @ 1.1-1.35 μm
      I_c ⇨ 396-492 A/cm
    - SP continuous process (MOCVD YBCO/IBAD MgO)
      - Short lengths > 400 A/cm ($J_e \sim 4.4 \times 10^4$ A/cm)
      - 10 m @ 139 A/cm
      - 25 m @ 142 A/cm

Scoring criterion - Performance

- Modify IBAD assist gun to expand deposition zone length to improve process efficiency. *Goal: Double length of deposition zone*.
- Improve IBAD MgO texture by reducing divergence of SuperPower's ion-assist gun. *Goal: Routinely obtain $\Delta\Phi \leq 5°$ FWHM.*
  - Improved cooling of the tapes during deposition allowed both goals to be satisfied via single approach
  - ✓ Deposition zone length increased from 8 to 16 cm
  - ✓ Texture for reel-to-reel processing improved such that $\Delta\Phi = 5°$
  - The processing window for which $\Delta\Phi \leq 8°$ was increased by ~ 2X - implies better yield/quality during manufacturing

Scoring criterion - Performance

- Expand upon our data of how YBCO superconducting properties (T_c, J_c) are affected by transition metal impurities. *Goal: Determine tolerance limits for substrate elements in YBCO films.*
  - Found less sensitivity to Ni impurity levels for T_c than for J_c
  - ✓ Tolerance limit for Ni in YBCO before rapid drop of J_c is ≤ 0.25 atomic %

Scoring criterion - Research Integration

- LANL-SuperPower CRADA two year renewal just completed
- Our primary research integration activity this year was the transfer of IBAD MgO technology to SuperPower. This was accomplished by sample and information exchanges, equipment loans, 3 on-site visits by LANL staff at their plant and 2 visits by SuperPower staff at LANL.
- SuperPower supplied LANL with ~ 100 m of electropolished C-276 alloy for IBAD texturing experiments with moving substrates.
- SuperPower supplied LANL with many meters of high-quality IBAD MgO with same texture for PLD YBCO single and multi-layer optimization experiments
- SuperPower loaned LANL one of their RF ion assist guns - integral to:
  - optimization of IBAD texturing deposition parameters on moving tapes
  - optimizing performance of SuperPower pilot system gun
- SuperPower provided 9 m lengths of 100 A/cm coated conductor to LANL to be fabricated into a coil

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Scoring criterion - Research integration

- Joint collaborations with: Ames, Argonne, Brookhaven, Oak Ridge and Sandia National Laboratories, Wright Patterson AFB, Universities of Augsburg, Cambridge, Stanford and Wisconsin

- Publications: 1 book chapter, 26 journal articles (3 in press, 12 with universities, national laboratories and companies) (List included at end of the handout with external collaborators highlighted)

- Patents issued: 6 (1 joint patent with Stanford University)
  - 6,921,741, "Substrate structure for growth of highly oriented and/or epitaxial layers thereon"
  - 6,899,928, "Dual ion beam assisted deposition of biaxially textured template layers"
  - 6,884,527, "Biaxially textured composite substrates"
  - 6,843,898, "High temperature superconducting composite conductors"
  - 6,800,591, "Buffer layers on metal alloy substrates for superconducting tapes"
  - 6,756,139, "Buffer layers on metal alloy substrates for superconducting tapes"

- Patent applications: 10 (1 joint application with ORNL), Patent disclosures: 1

Scoring criterion - FY2006 plans

- Continue to address strategies of mutual interest with SuperPower for increasing performance and reducing costs

  Goal: Transfer IBAD cooling technology to SuperPower

  Goal: Determine which RE(Zr, Hf)O buffer is most amenable to SuperPower's processing requirements & transfer to SuperPower.

  Goal: Transfer multilayer architecture to SuperPower - Reproducible 500 A/cm-width in 2 μm.

- Continue investigation into effects of transition metal impurities on YBCO (T_c, J_c) *Goal: Determine tolerance limit for Cr in YBCO films and refine data for Ni.*

- Study ion-damage anisotropy of MgO for Ar^+ energies comparable to those used in IBAD processing *Goal: Develop a model that will allow us to further refine IBAD deposition parameters and texture.*